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Mechanical Metallurgy of Plutonium Metal, Alloys, and Allotropes

Michael F. Stevens

May 5, 2020
Actinide Science and Technology Lecture Series

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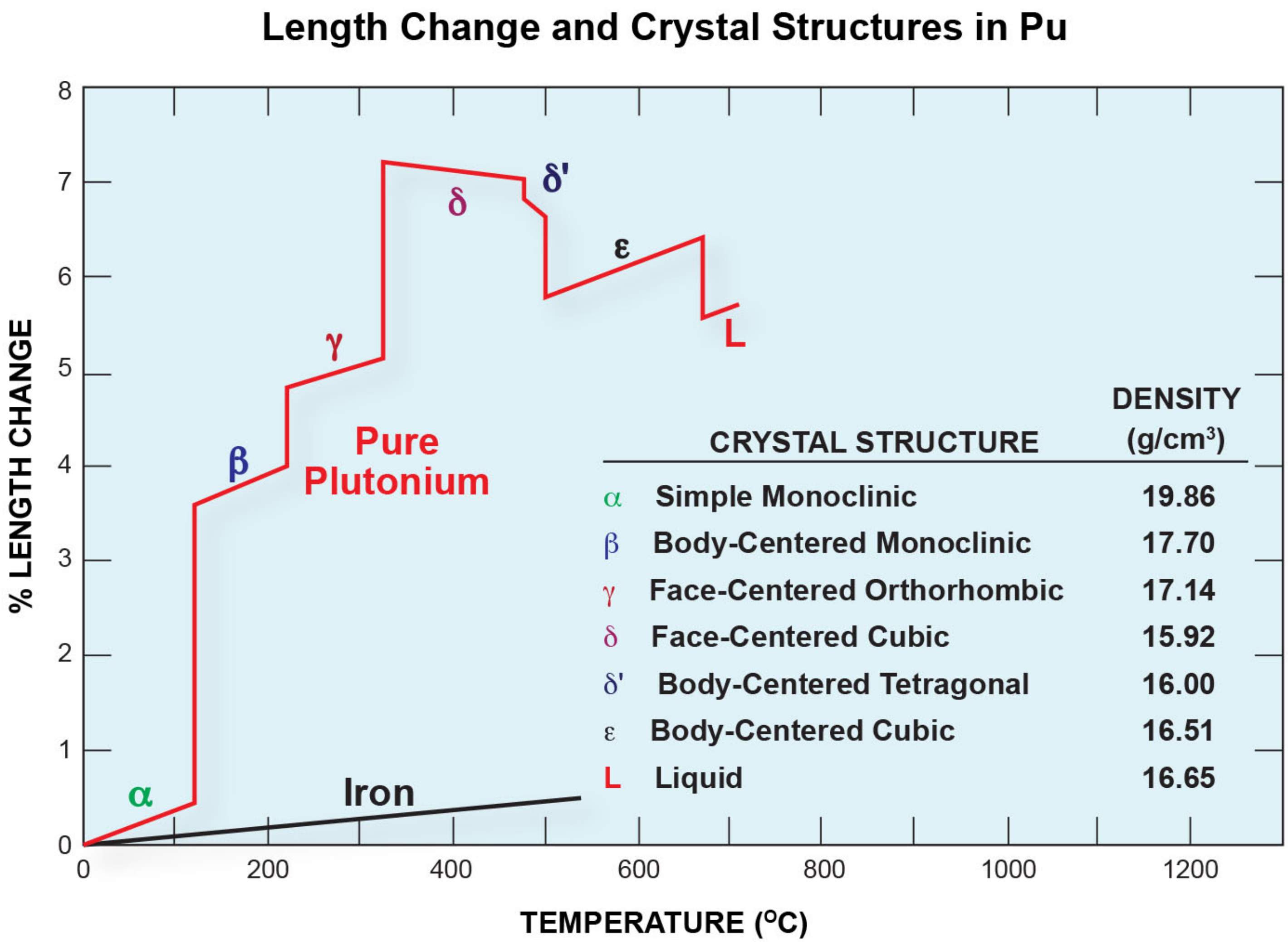
LA-UR-19-31449

“Mechanical Metallurgy” of Plutonium

- Talk will highlight the mechanical properties of plutonium, but with some discussion of the governing (and often unusual!) underlying metallurgical fundamentals
- In the interest of time, this talk will focus on unalloyed plutonium metal, its temperature allotropes, and plutonium alloyed to stabilize the delta phase
- Most of the featured work is taken from the literature; references are provided for both attribution and further reading.

Plutonium is both Complex and Unstable to changes to environment – basis is unique electronic structure!

See Hecker, S. S., *Los Alamos Science*, 2000, **26**, 290-335



CRYSTAL STRUCTURE		DENSITY (g/cm³)
α	Simple Monoclinic	19.86
β	Body-Centered Monoclinic	17.70
γ	Face-Centered Orthorhombic	17.14
δ	Face-Centered Cubic	15.92
δ'	Body-Centered Tetragonal	16.00
ε	Body-Centered Cubic	16.51
L	Liquid	16.65

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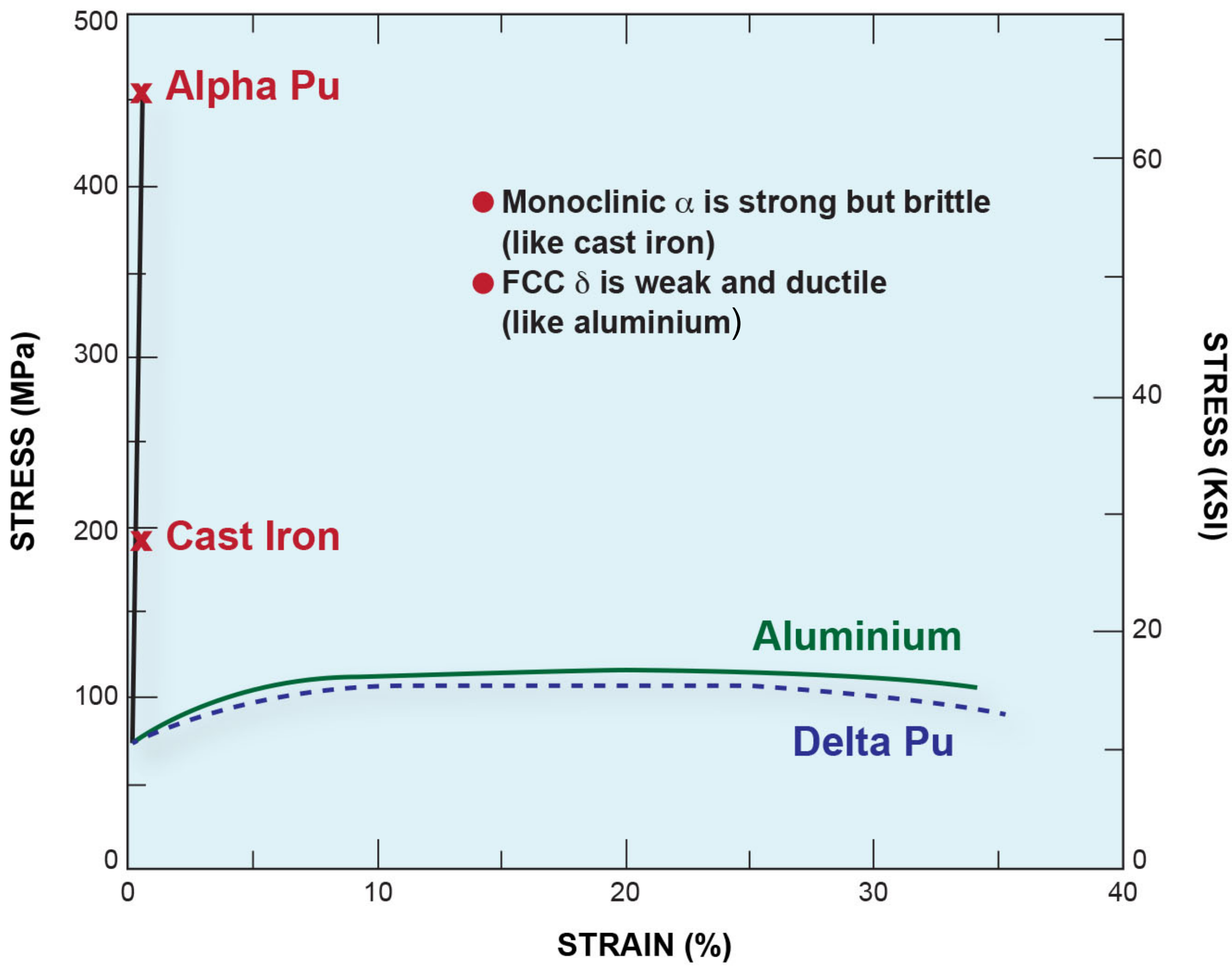
Tensile Properties of α and δ Pu

Mechanical Properties are Very Sensitive to Crystal Structure



From Hecker, S. S.; Stevens, M. F. *Los Alamos Science*, 2000, **26**, 336-355

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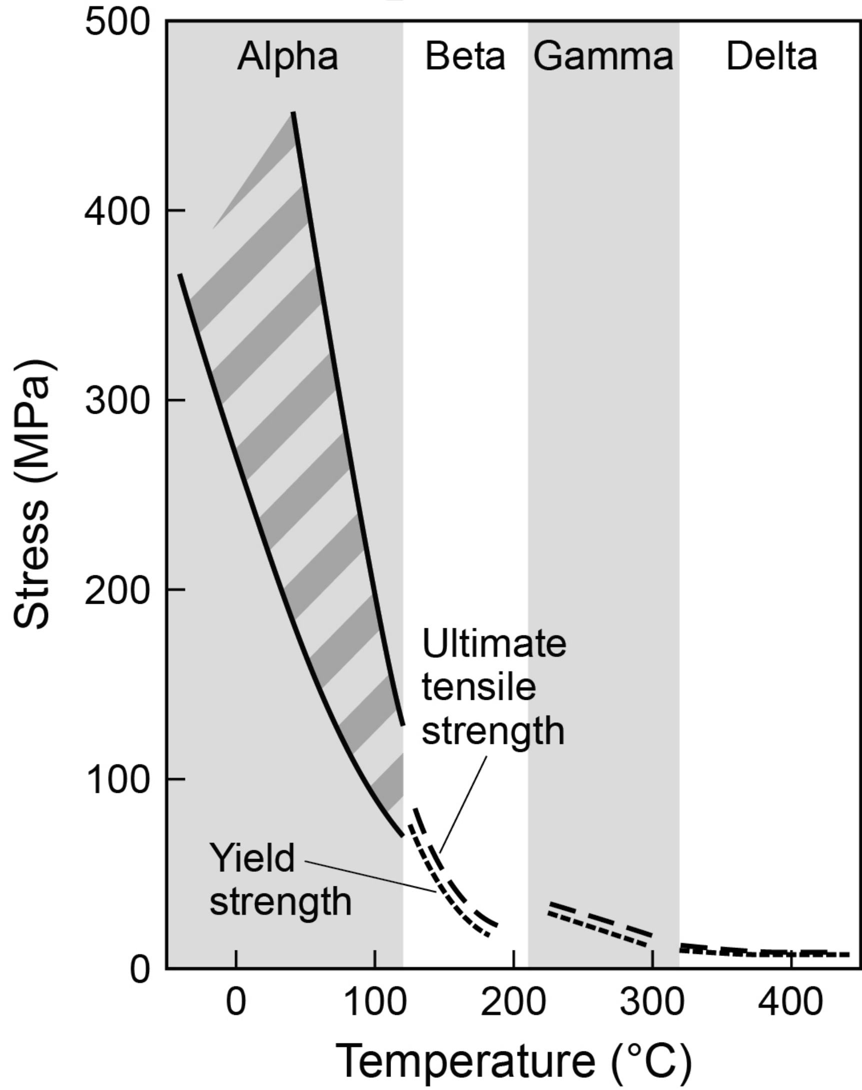
Plutonium Metal - Allotropes

Tension

From Gardner, H. R.; Mann, I. B. Mechanical Property and Formability Studies on Unalloyed Plutonium. In Plutonium 1960: The Proceedings of the Second International Conference on Plutonium Metallurgy, Grenoble, France, April 19–22, 1960; Grison, E., Lord, W. B. H., Fowler, R., Eds.; Cleaver-Hume Press: London; pp 513–570.



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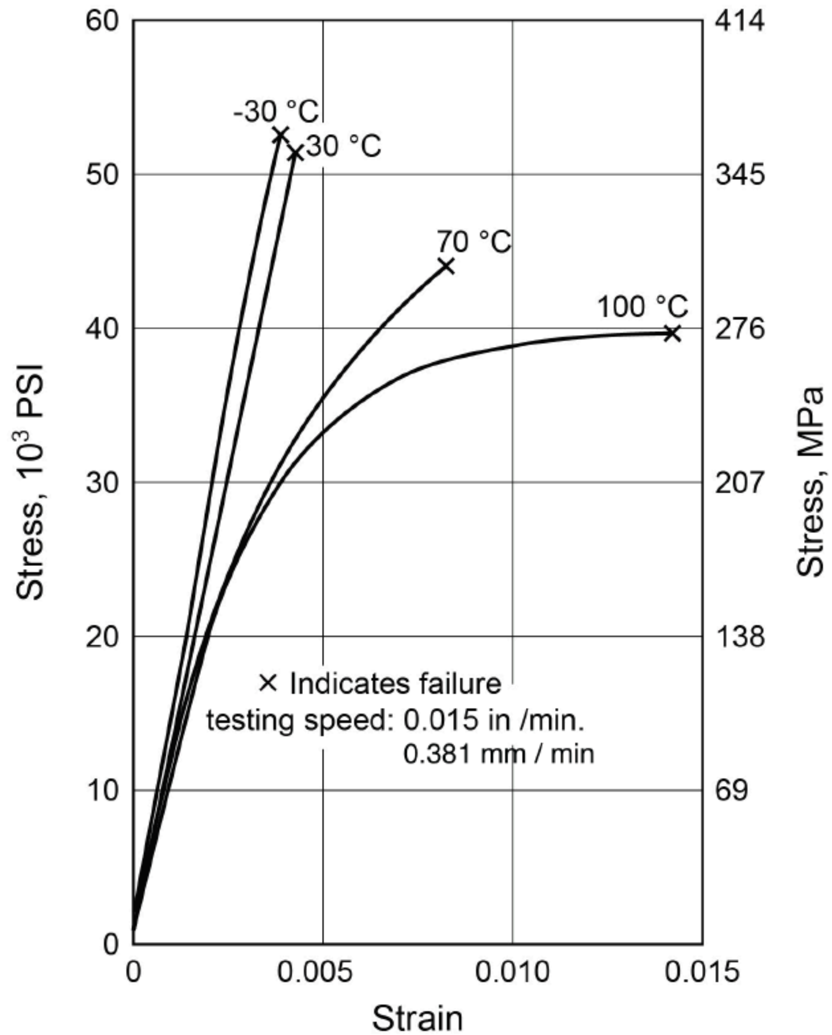
General observations of α–phase deformation

- Negligible macroscopic ductility in tension generally attributable to presence of microcracks (stress concentrators) resulting from $\beta\rightarrow\alpha$ volume collapse during solidification
- Microscopic fracture surfaces reveal extensive, but shallow microvoid coalescence; stress induced transformation conjectured.
- Exhibits *decreased* hardness with decreasing grain size
- Anomalously high homologous temperature ~ 0.53[1] at RT allows for a variety of non-conserving deformation mechanisms, especially in fine grained samples; enhanced ductility results
- Extruded, fine-grained alpha displays enhanced tensile ductility; Merz[2] hypothesized that fine grain structure (grain boundary sliding) and absence of microcracks responsible.

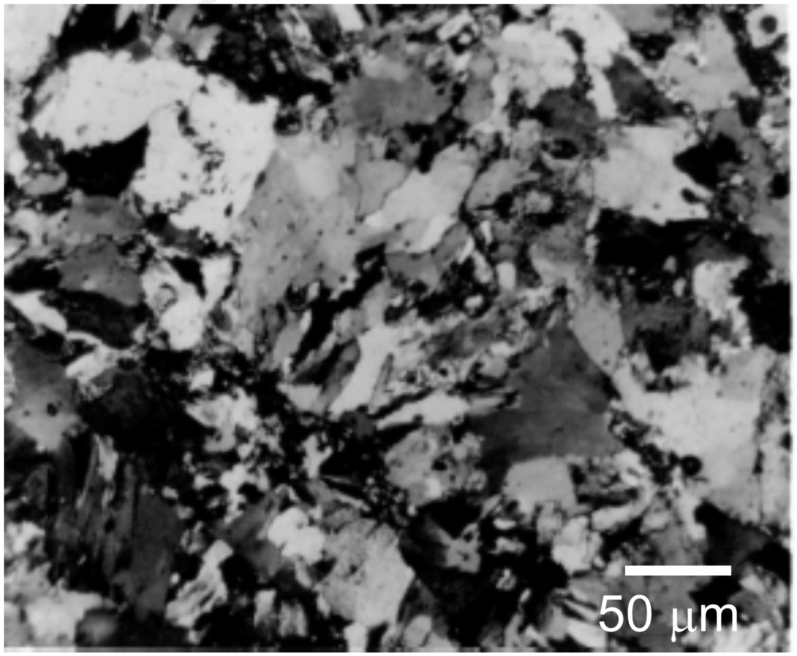
[1]Nelson, R. D. Bierlein, T. K., Bowman, F. E., USAEC Report BNWL-32 (1965).
[2]Merz, M. D. *J. Nucl. Mater.* 1971, 41, 348–350.

Tensile Behavior of α - Plutonium

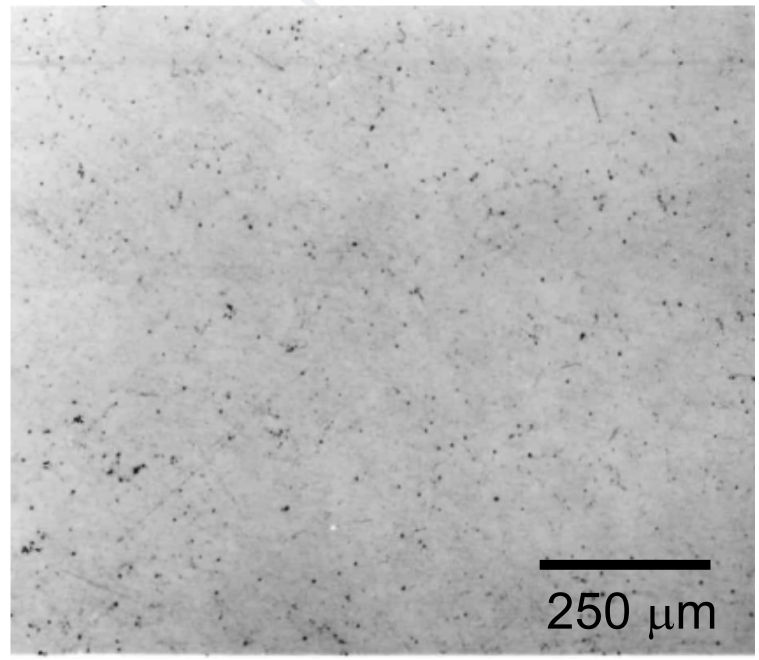


From Gardner, H. R., Hanford Atomic Products Operation, General Electric Co., Unpublished Data, April 1962.

Alpha Pu microstructure



Typical Grain Structure of As-Cast Alpha Pu
Grain Size – 47 μm
Density – 19.54 g/cc



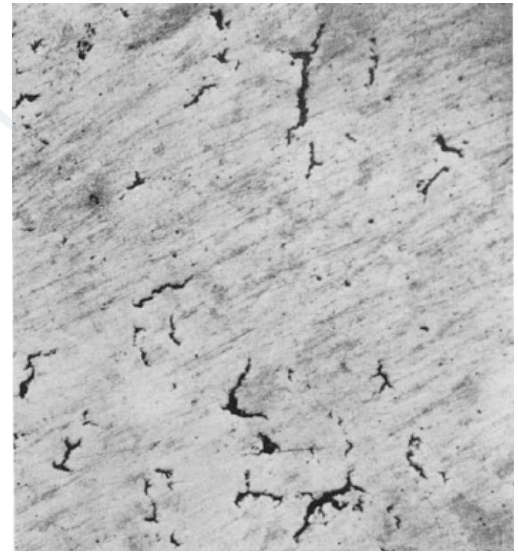
As polished surface of As-Cast Alpha Pu
Density – 19.56 g/cc
Microcracks and voids evident

From Gardner, H. R.; Mann, I. B. Mechanical Property and Formability Studies on Unalloyed Plutonium. In Plutonium 1960: The Proceedings of the Second International Conference on Plutonium Metallurgy, Grenoble, France, April 19–22, 1960; Grison, E., Lord, W. B. H., Fowler, R., Eds.; Cleaver-Hume Press: London; pp 513–570.

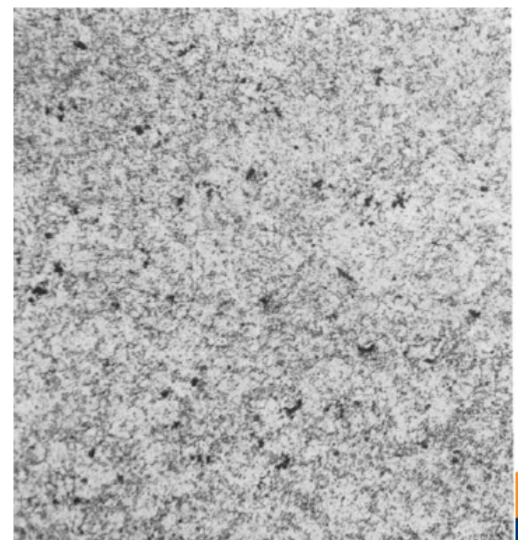
Microcracking in α–phase Plutonium – Impurity and Processing Effects

- Impurities known to reduce microcracking through secondary phase formation (e.g. Pu_6Fe)[1]
- Harbur, *et al.*[2] found that chill casting could suppress microcracks through optimization of transformation front.
- Extrusion of α–phase Pu also results in enhanced ductility[3] due to closure of microcracks and grain boundary sliding

ρ = 19.64 g/cc
Mold T 0℃



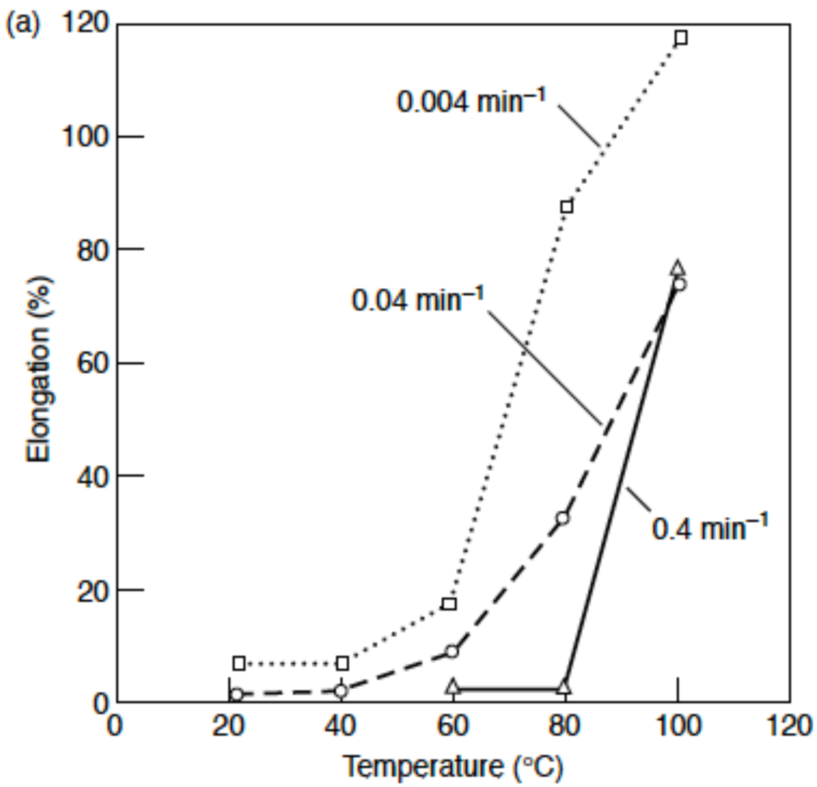
ρ = 19.79 g/cc
Mold T -80℃



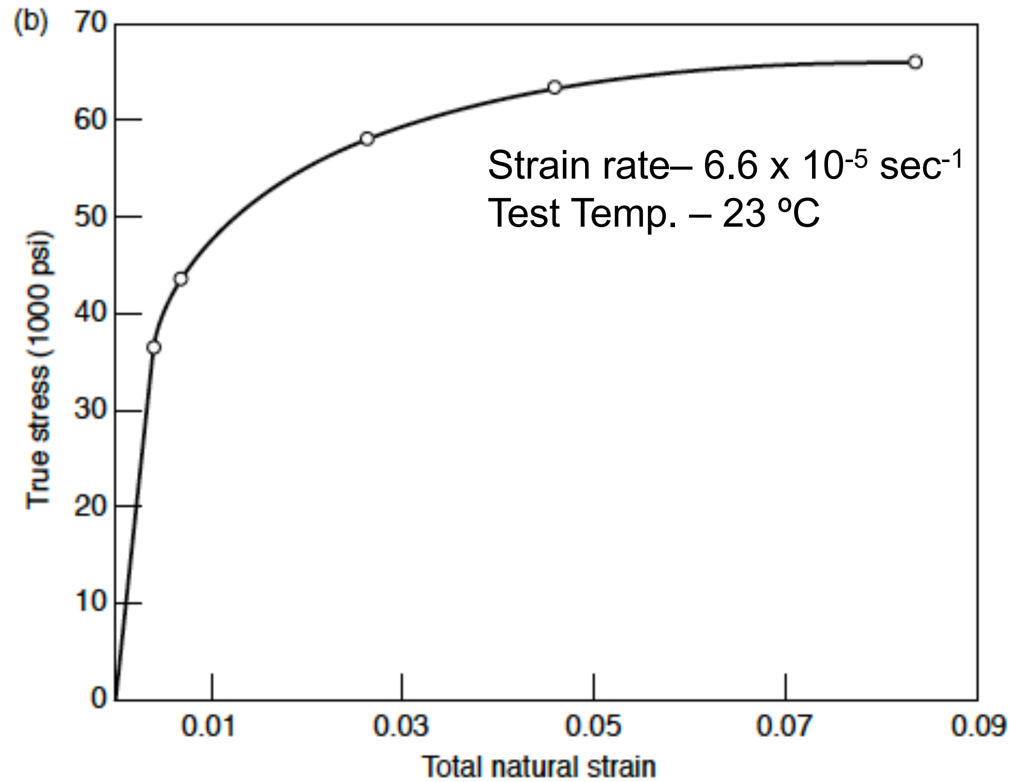
[1]Gardner, H. R.; Mann, I. B. Mechanical Property and Formability Studies on Unalloyed Plutonium. In *Plutonium 1960: The Proceedings of the Second International Conference on Plutonium Metallurgy,* Grenoble, France, April 19–22, 1960.
[2]Harbur, D. R.; Romero, J. W.; Anderson, J. W.; Maraman, W. J. *J. Nucl. Mater.* 1968, 25, 160–165.
[3]Merz, M. D. *J. Nucl. Mater.* 1971, 41, 348–350.

Enhanced ductility in extruded, fine grain α – Pu

Typical grain size – 1 to 3 μm



From Merz, M. D.; Allen, R. P. *J. Nucl. Mat.* 1973, 46, 110-112.



From Merz, M. D. *J. Nucl. Mat.* 1971, 41, 348-350.

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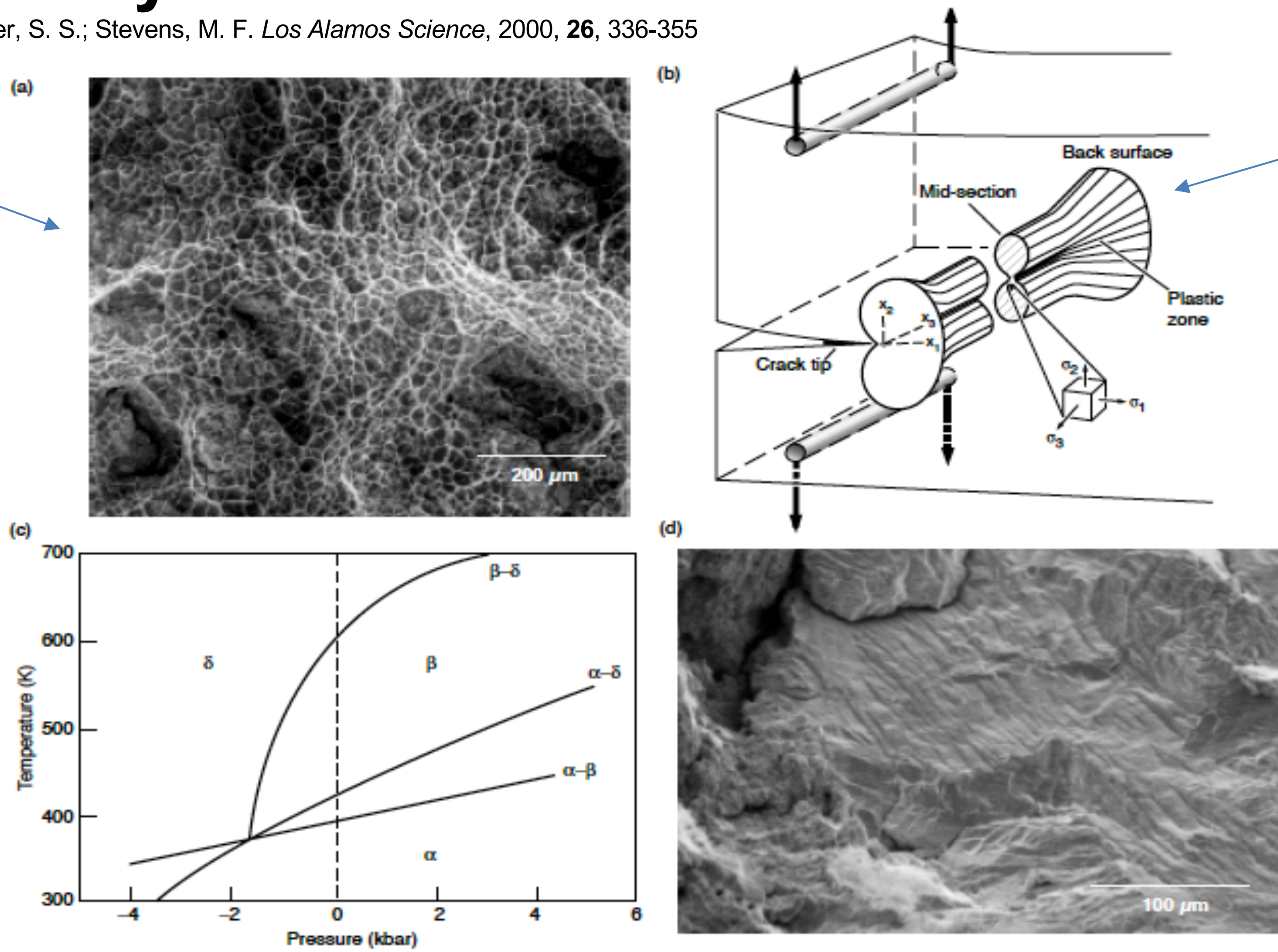
Evidence for $\alpha \rightarrow \delta$ Transformation under Hydrostatic Tension

From Hecker, S. S.; Stevens, M. F. *Los Alamos Science*, 2000, **26**, 336-355

Tensile fracture surface

Schematic showing triaxial stress zone at loaded crack tip



Torsion fracture surface

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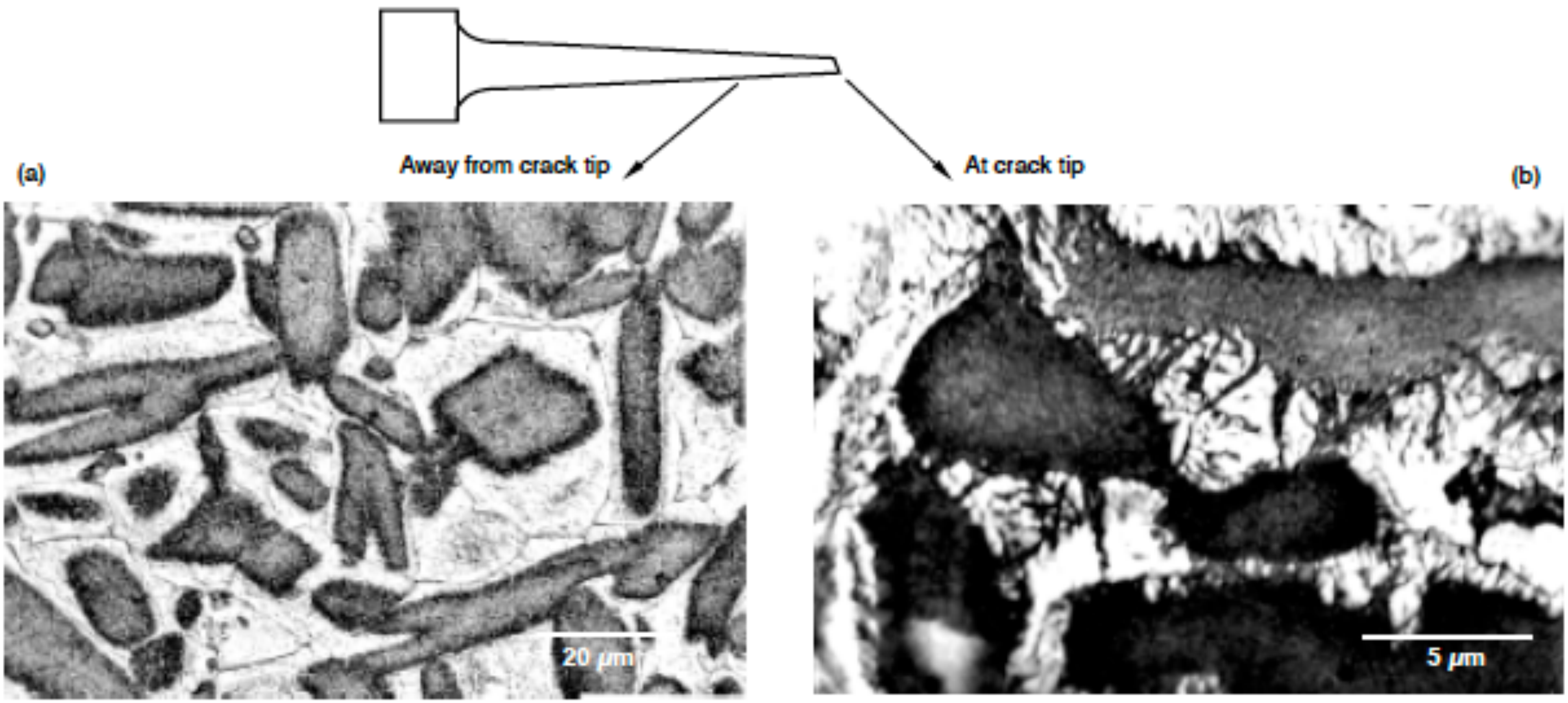
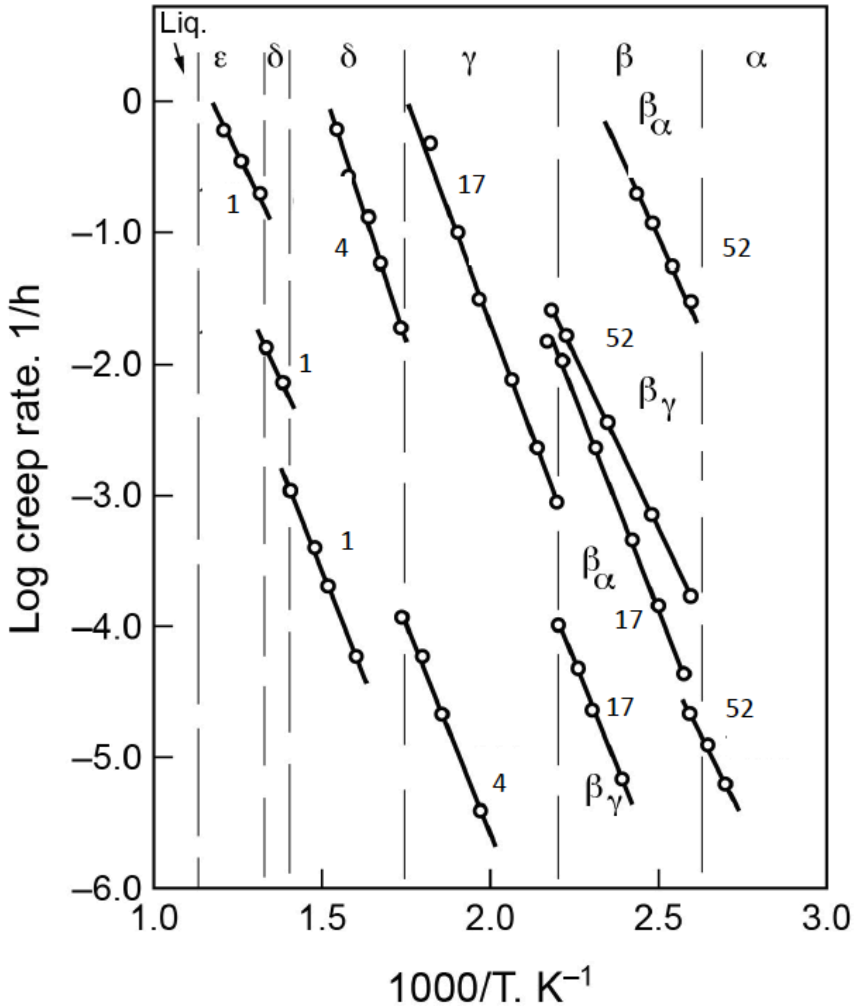
Further Evidence for $\alpha \rightarrow \delta$ Transformation during Hydrostatic Tension



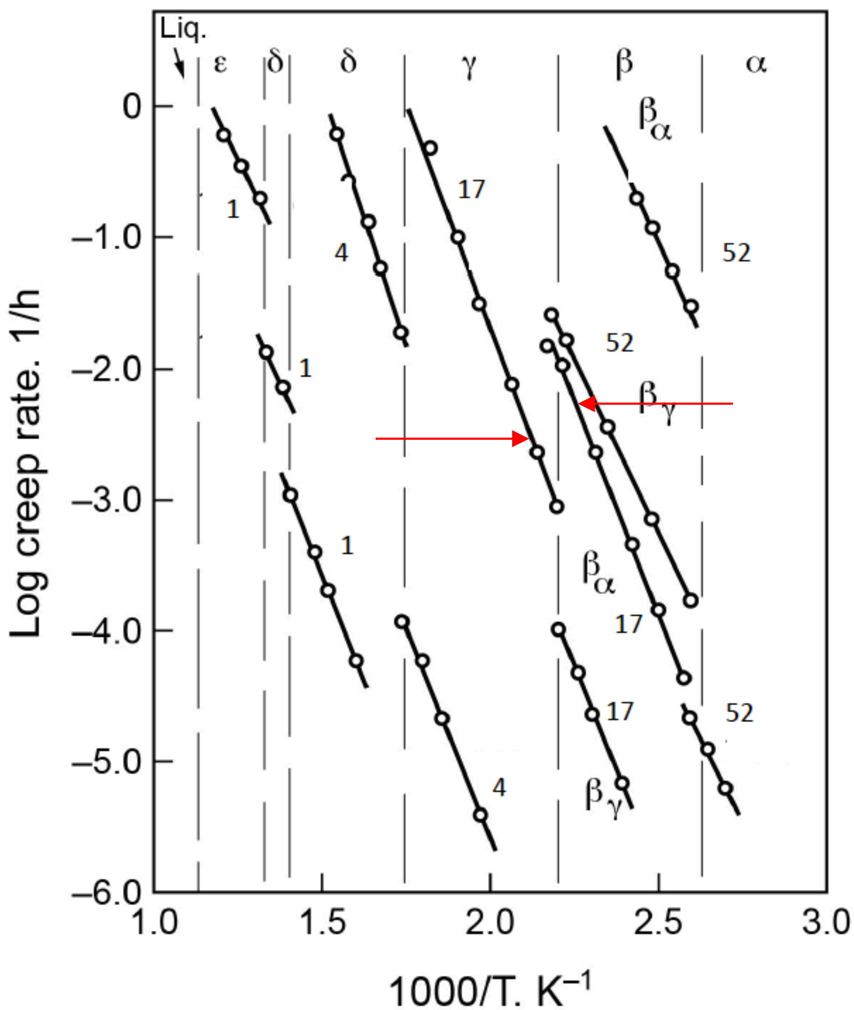
From Hecker, S. S.; Stevens, M. F. *Los Alamos Science*, 2000, **26**, 336-355

Creep Deformation of Plutonium



From Nelson, R. D. Steady-State Creep of Plutonium. in *Plutonium 1965: Proceedings of the Third International Conference on Plutonium: London 1965*; Kay, A. E., Waldron, M. B., Eds.; Chapman & Hall: London, 1967; pp 564–570.

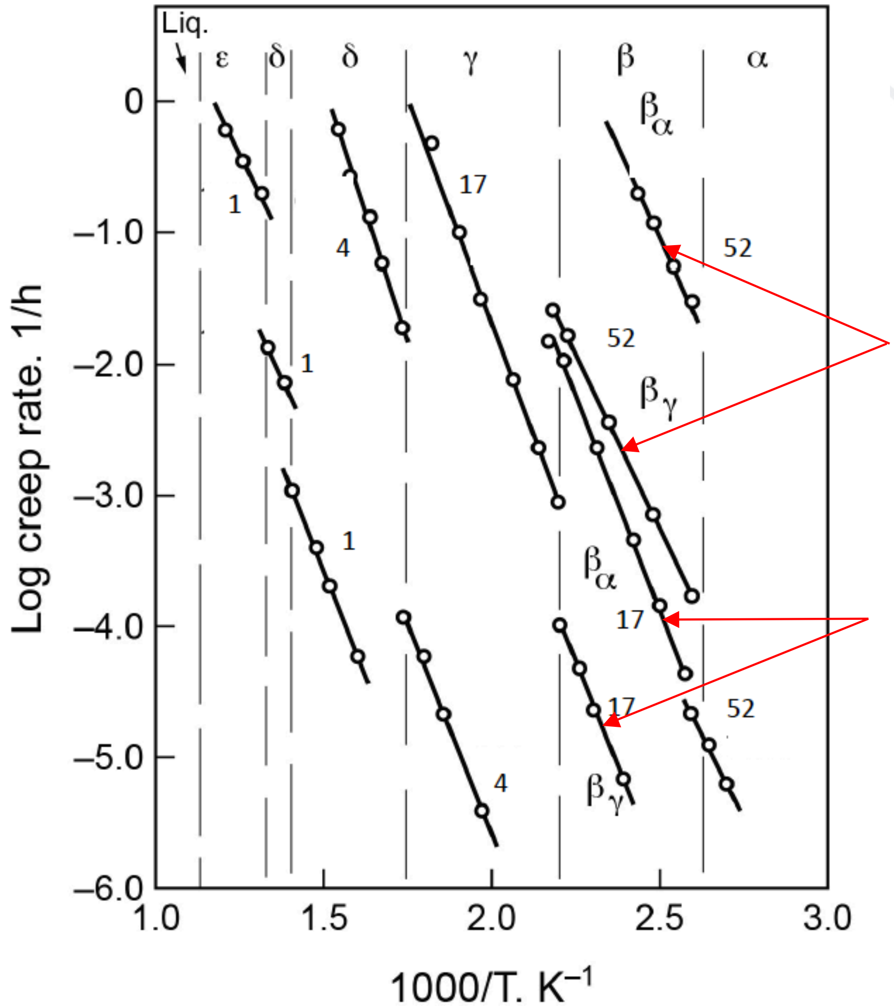
Creep Deformation of Plutonium



Lower temp allotrope has higher creep rate at equiv. stress level!

From Nelson, R. D. Steady-State Creep of Plutonium. in *Plutonium 1965: Proceedings of the Third International Conference on Plutonium: London 1965*; Kay, A. E., Waldron, M. B., Eds.; Chapman & Hall: London, 1967; pp 564–570.

Creep Deformation of Plutonium



β_γ always has a lower creep rate than β_α

From Nelson, R. D. Steady-State Creep of Plutonium. in *Plutonium 1965: Proceedings of the Third International Conference on Plutonium: London 1965*; Kay, A. E., Waldron, M. B., Eds.; Chapman & Hall: London, 1967; pp 564–570.

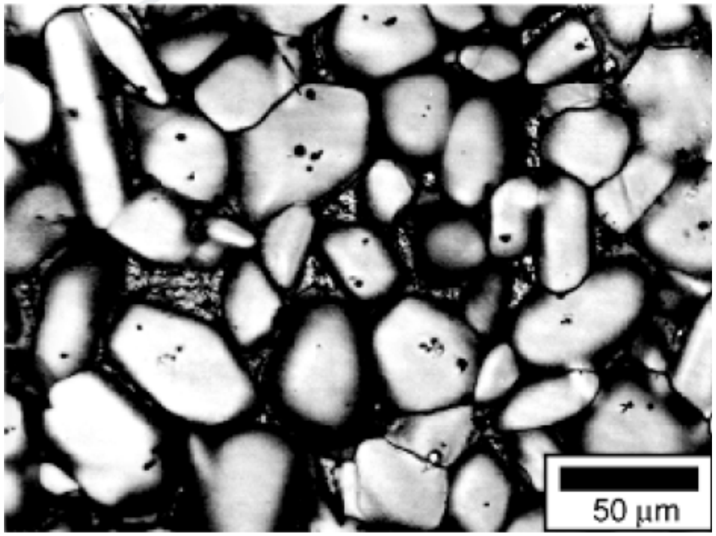
δ–phase Plutonium

- Lowest density of the solid state allotropes
- Face-centered cubic crystal structure
- Soft, ductile; comparable to 1100 Al
- Stabilized to RT by several, usually trivalent alloying elements – Al, Si, Ga, Ce, Am
- High elastic anisotropy
- Anomalous thermal expansion

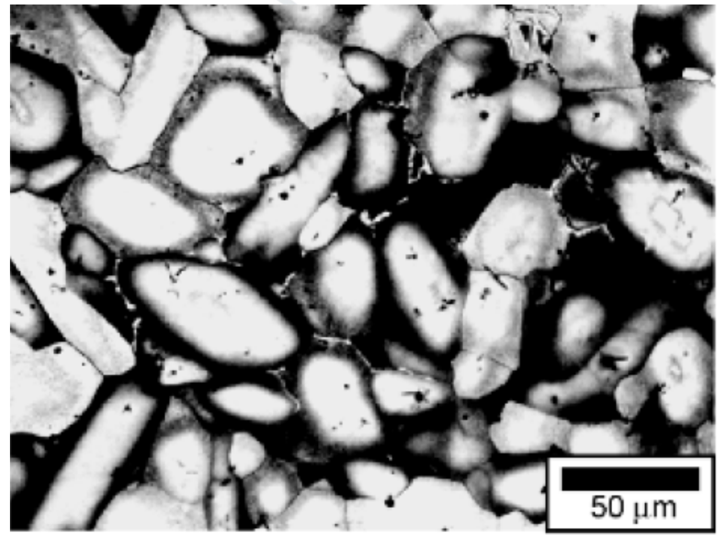
Small Ga additions stabilize the δ–phase (3.4 at. % Ga)

Mitchell, J. N., Gibbs, F. E., Zocco, T. G., and Pereyra, R. A. (2001) Metall. Mater. Trans. A, 32(3A), 649–59.

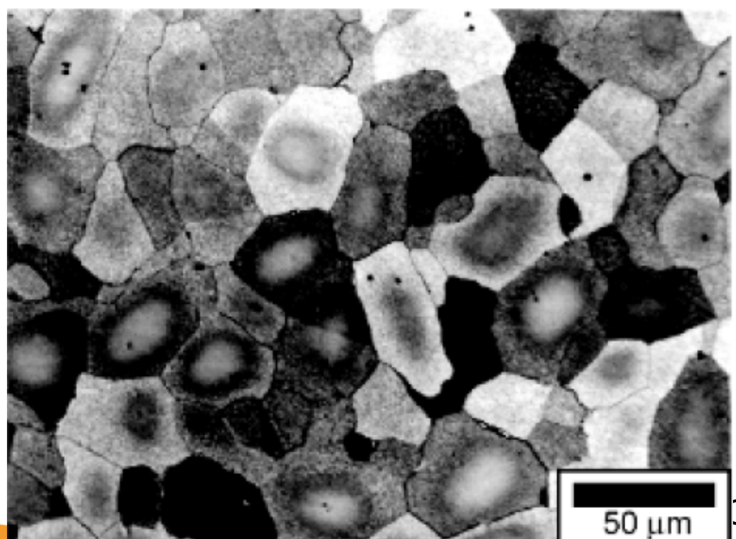
As-cast



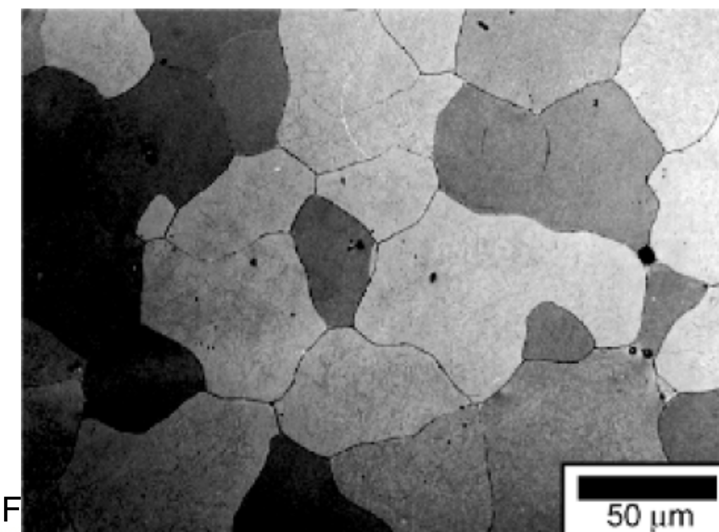
460ºC – 2 hrs



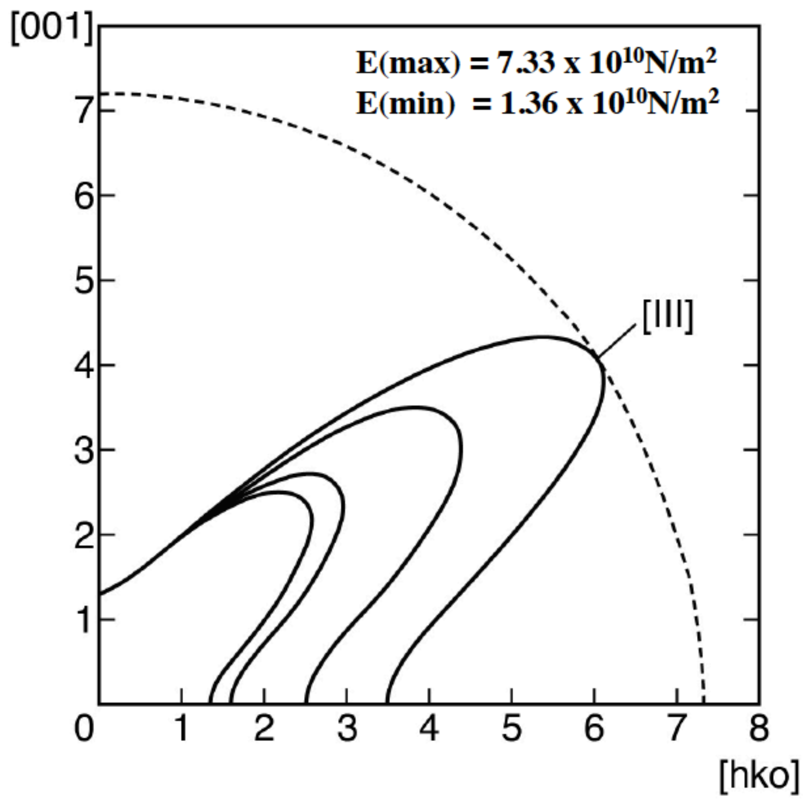
460ºC – 20 hrs



460ºC – 720 hrs



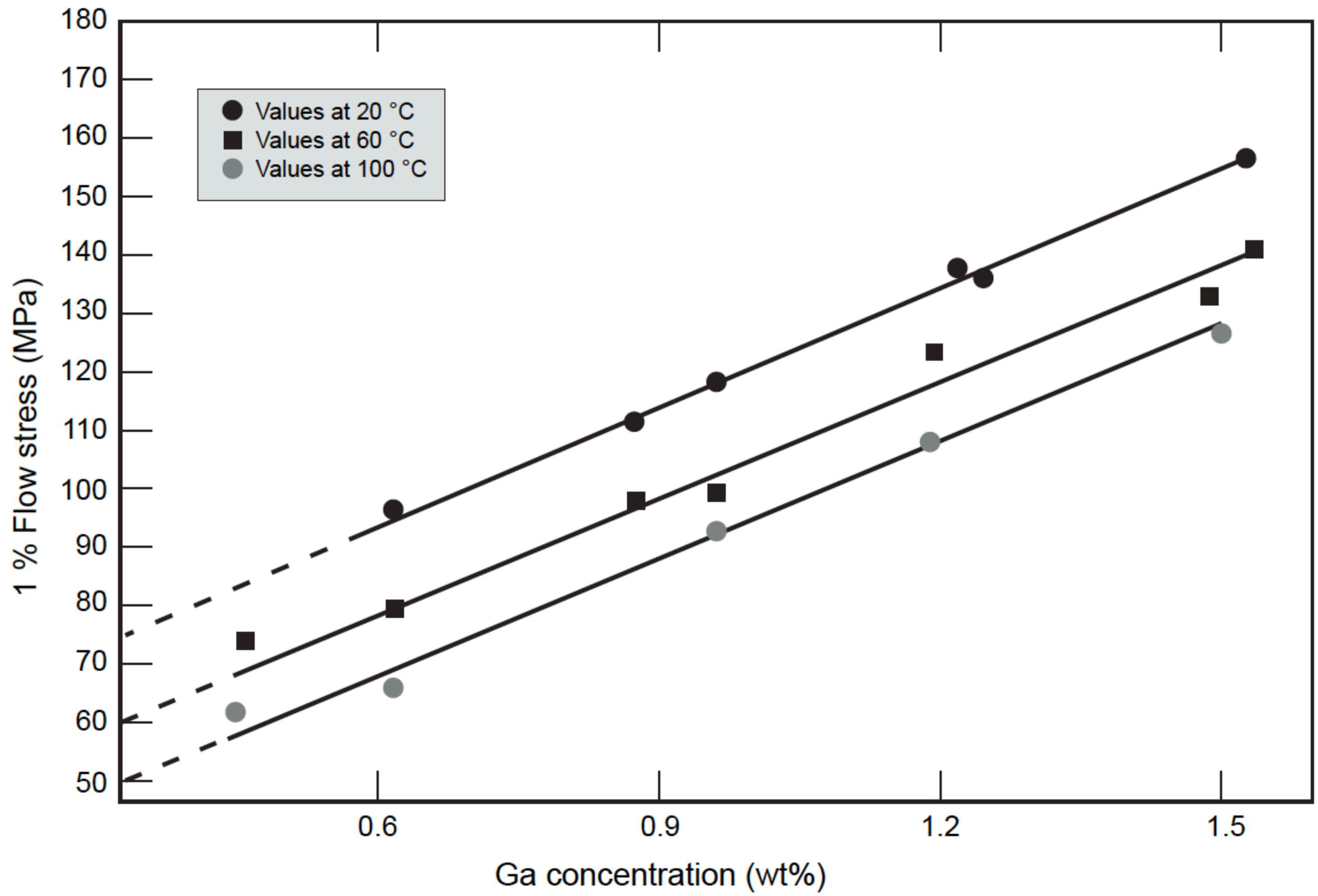
δ–Phase Plutonium Elastic Properties



Polar plot of Youngs modulus vs. lattice direction in a Pu – 1 wt. % Ga single crystal

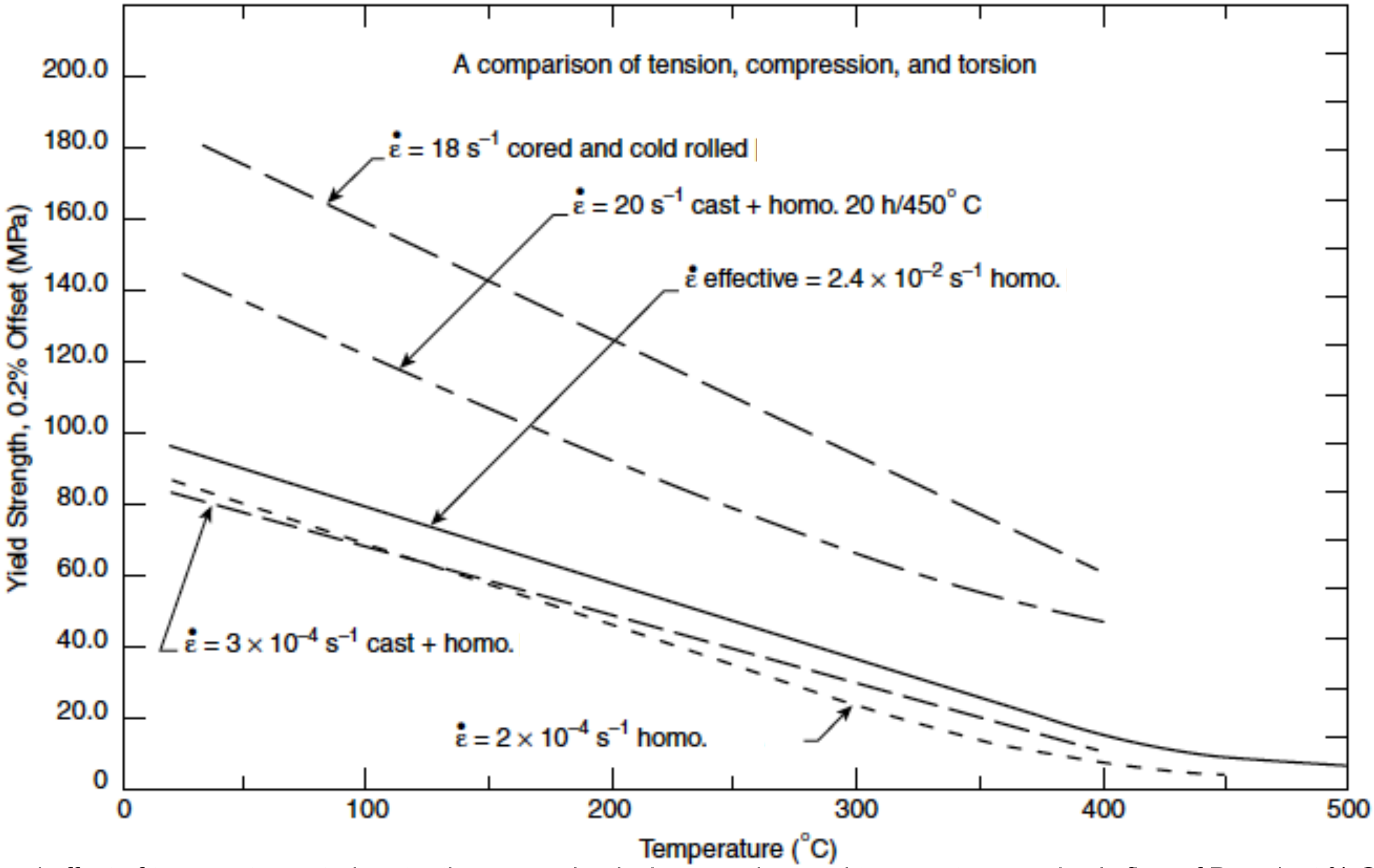
From Ledbetter, H. M.; Moment, R. L. *Acta Metall.* 1976, 24, 891–899.

Ga effect on Flow Stress in δ -Pu



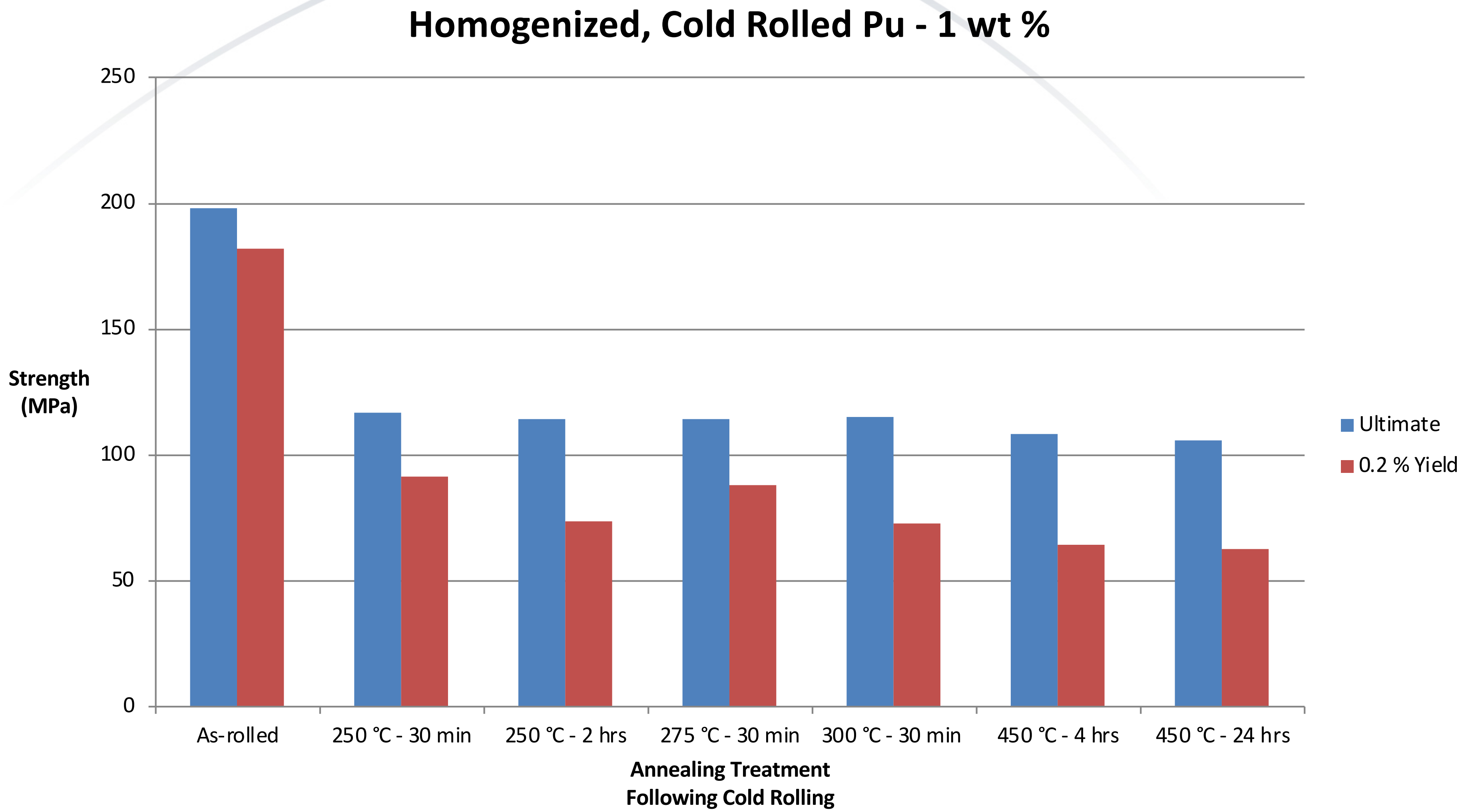
From Miller, D. C.; White, J. S. *J. Nucl. Mat.* 1965, 17, 54-59.

Recent Studies Expanded on Pu-Ga Alloy Behavior



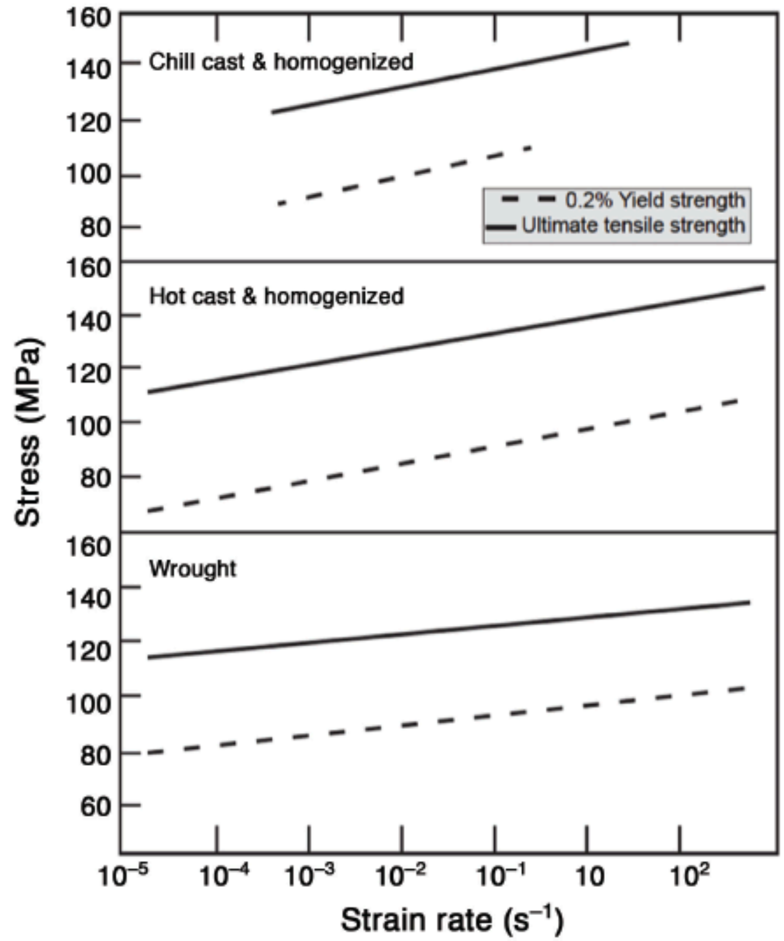
Combined effect of temperature, strain rate, thermomechanical processing, and temperature on plastic flow of Pu – 1 wt.% Ga alloys. From Robbins, *J. Nucl. Mater.* **2004**, 324, 125-133.

Annealing Response of δ–phase Pu Recrystallization Kinetics



From Gill, S. M.; *BNWL-SA-380, Conf-651119-4, October 22, 1965*

Tensile Flow of Pu-1 wt% Ga Alloy vs Strain Rate and Thermomechanical Treatment



From Hecker, S. S.; Morgan, J. R. Effect of Strain Rate on the Tensile Properties of α- and δ-Stabilized Plutonium, in *Plutonium 1975 and Other Actinides: Proceedings of the Conference in Baden Baden, September 10–13, 1975: 5th International Conference on Plutonium and Other Actinides*; Blank, H., Lindner, R., Eds.; North-Holland Publishing Company: Amsterdam, 1976; p 703.

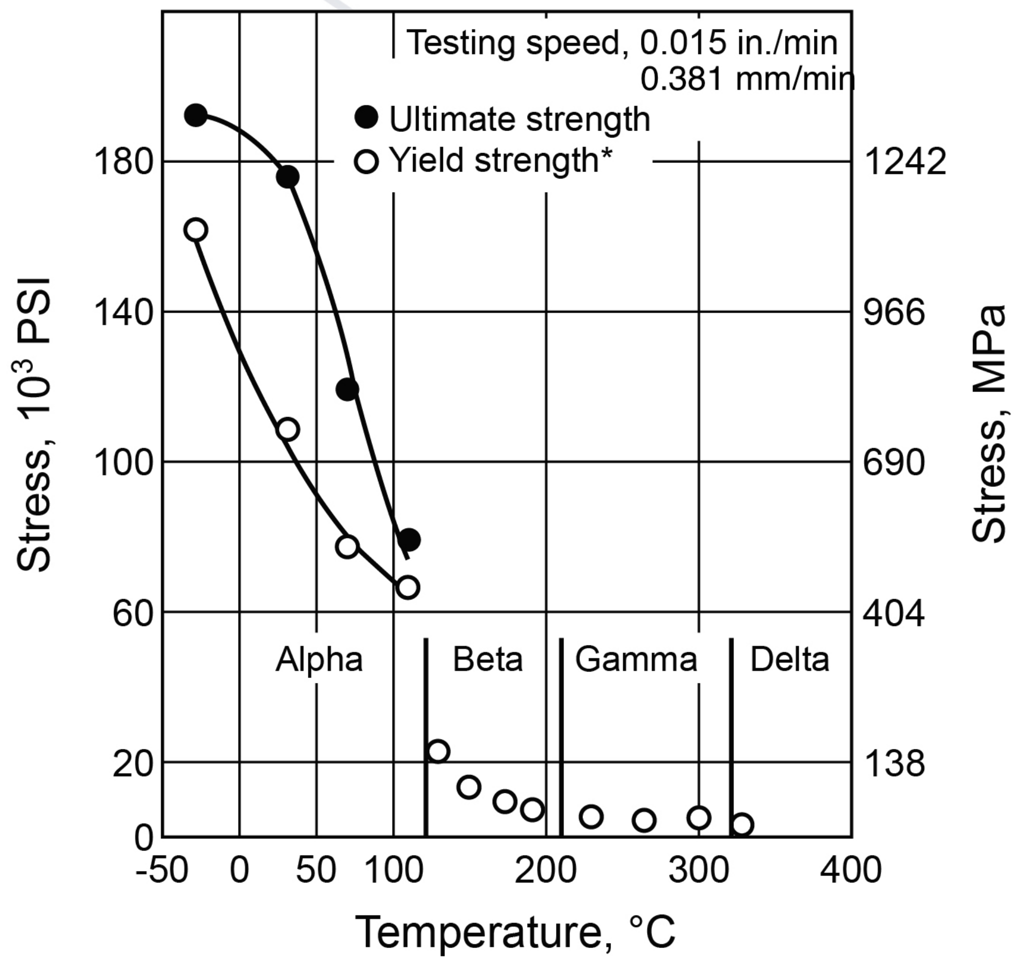
Takeaways

- Mechanical properties of plutonium are highly dependent on crystal structure, processing history, and alloy content
- Pure plutonium mechanical properties are strongly influenced by the low-symmetry crystal structure, but also defect content, homologous temperature, processing history, and perhaps phase stability
- Stabilized delta phase plutonium is well adapted for engineering applications

Plutonium Metal - Allotropes

Compression

From Gardner, H. R.; Mann, I. B. Mechanical Property and Formability Studies on Unalloyed Plutonium. In Plutonium 1960: The Proceedings of the Second International Conference on Plutonium Metallurgy, Grenoble, France, April 19–22, 1960; Grison, E., Lord, W. B. H., Fowler, R., Eds.; Cleaver-Hume Press: London; pp 513–570.



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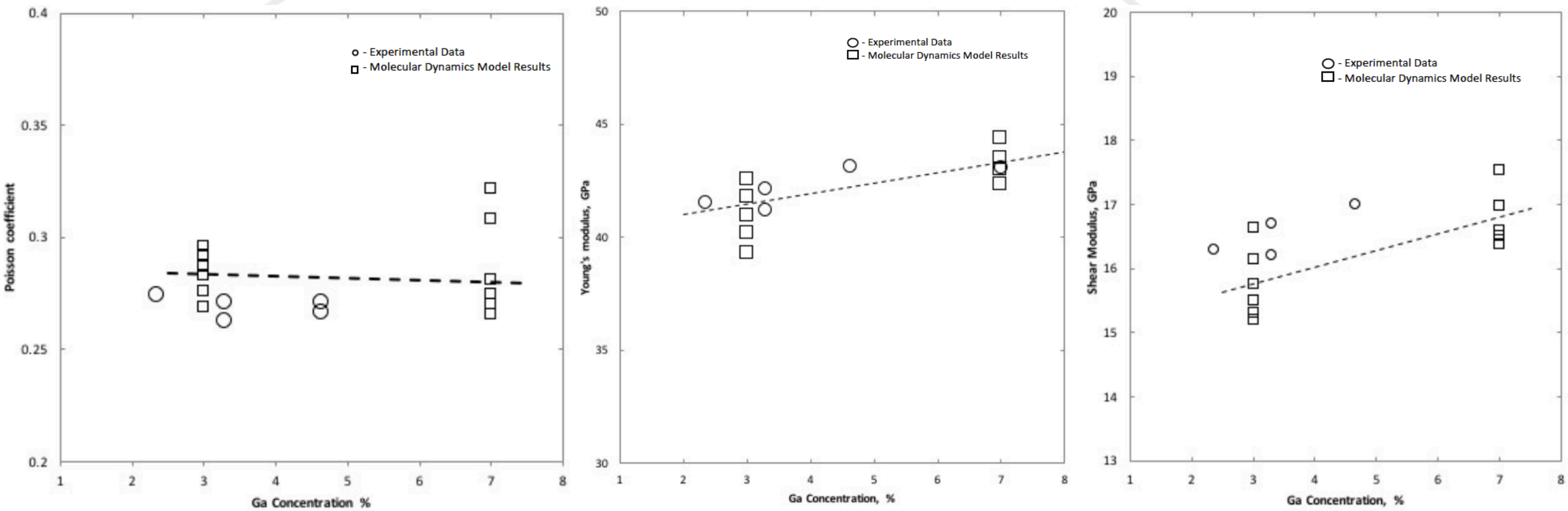
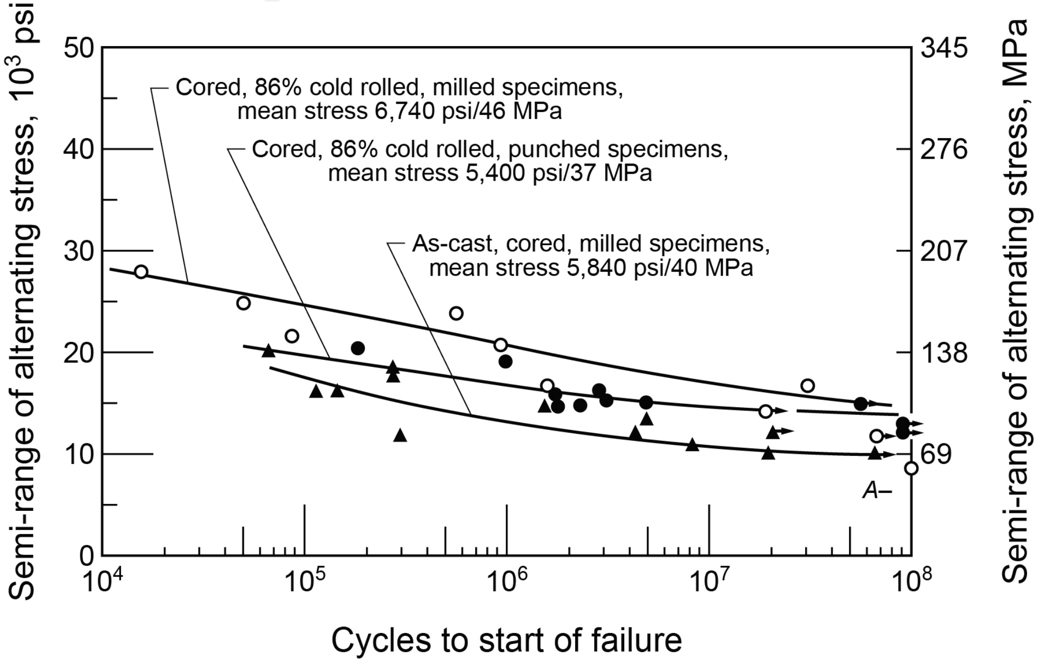
Classical Molecular Dynamics Predictions of Elastic Properties

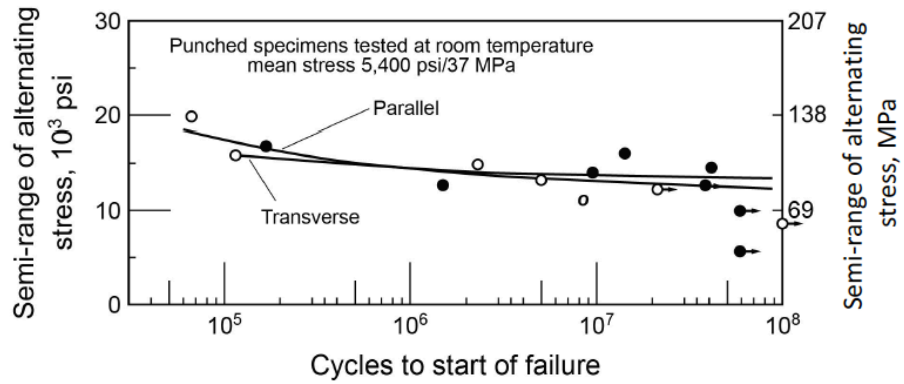


- Accurate MEAM potentials give good agreement of elastic properties predictions with empirical data
- Results extrapolated to predict effect of age induced atomic scale defects (i.e. vacancies, He atoms) on elastic and plastic properties

From Dremov, V. V.; Karavaev, A. V.; Sapozhnikov, F. A.; Vorobyova, M. A.; Preston, D. L.; Zocher, M. A. *J. Nucl. Mater.* 2011, 414, 471–478.
Experimental data from A. Migliori et al., *J. Alloys Compd*. 444–445 (2007) 133–137.

Fatigue behavior of δ-Stabilized Plutonium





From Gardner, H. R. Fatigue Behavior of Plutonium-1 wt % Gallium δ-Stabilized Plutonium.
In *Plutonium 1965: Proceedings of the Third International Conference on Plutonium: London 1965*; Kay, A. E., Waldron, M. B., Eds.; Chapman & Hall: London, 1967; pp 535–542.